Surface Composition Influence on Internal Gas Flow at Large Knudsen Numbers

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Abstract. An experimental study of the gas molecule conductivity for cylindrical channel with the surface prepared by metal spray technique has been carried out. The dramatically change of a gas conductivity value is observed at the process of residual gas adsorption on a clean metal surface. Vast experimental material on molecular beam scattering and theoretical approach for description of a gas molecules scattering on the solid state surface are used for comparison and interpretation of the received experimental data. A satisfactory agreement between experimental results and numerical simulation of the effect is achieved.

INTRODUCTION

All experimental papers devoted to the problem of gas - surface interaction can be divided into two groups. The first one includes experiments with a solid state surface immersed into a gas at equilibrium state or at the process of relaxation to equilibrium. Such experiments give an information about the gas molecules scattering on the surface from macroscopic properties of a process, in which the reaction of all body on the gas - solids interface change is shown. The experiments with a metal wire cooling [1], plate vibration dumping [2], rotating disc [3] and so on were carried out in a rarefied gas using this approach.

The second group includes experiments on study a gas molecules reaction on the gas - solids interface change. That is, first, the experiments with molecular beam scattering on the solid surface. Such experiments use the incident beams which have the following properties: absence of molecule interaction in a beam, the directions of falling molecules should lay inside a small corporal corner, a molecule velocities are in the narrow interval. The incident angle and molecule velocity discrete change permits to build unequivocally an indicatrise of scattering. Then using some model of scattering one can receive gas - solids interface parameters.

The results of experiment presented in this paper are connected with a free molecular gas conductivity study of a cylindrical channel. This results characterize a gas molecule reaction on the channel surface condition change. A free molecular regime of a gas flow at the normal conditions correspondents to the flow without molecule interaction with continuous incident angles and a velocity spectrum corresponding to equilibrium Maxwell distribution function. From this point of view it would be reasonable to use for comparison and interpretation of the data obtained in this experiment the vast experimental material about a molecular beam scattering and created on their basis the formal approach for this process description [4].

EXPERIMENTAL BACKGROUND

The main idea of experiment is described earlier [1]. Pyrex glass cylindrical channel with a metal wire stretched alone its axis is situated in an ultra high vacuum system. The system is supplied with means of gas phase, surface

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Abstract

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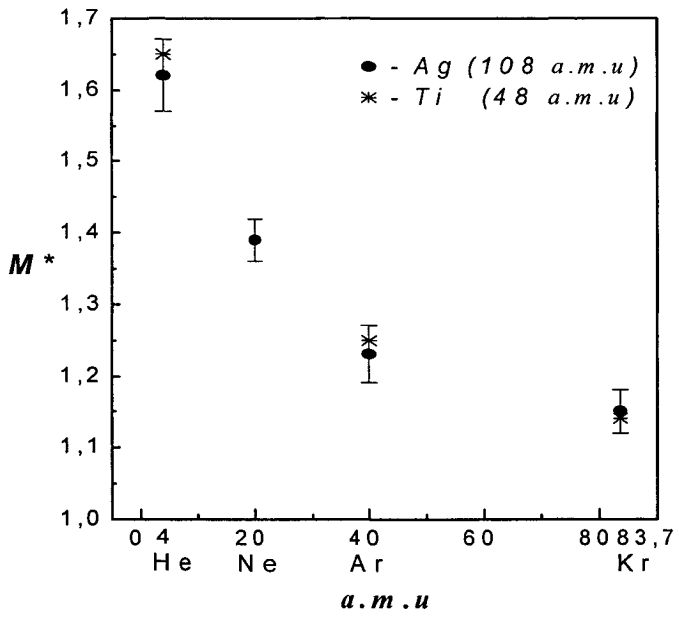
control and gas flow measuring system. The experimental procedure consists in a few stages. The first stage includes surface preparation process at ultra high vacuum conditions under residual pressure of about 10^{-10} *Pa*. At the next step free molecular conductivity measurement is realised for the channel with prepared surface and different gases. The method for unsteady flow first proposed by Knudsen and described in [5] has been used. Separate experiment was carried out for the surface covered by adsorbed gases. The chemical composition realized in this case is provided by means of long time exposition of the surface in residual gases.

EXPERIMENTAL RESULTS

Surface chemical composition influence on the gas scattering process and free molecular flow is studied for *He, Ne, Ar, Kr.* The chemical composition of the channel surface is changed from *Ag* atomic clean to completely adsorbed. Atomic clean surface prepared by spraying technique are used in the experiment. A non-dimensional relative gas conductivity of the channel M^* with *Ag* and *Ti* atomic clean surface as a function of a gas nature is presented at figure 1. The M^* value is standardized on the value of gas conductivity corresponding to completely diffuse interaction of the molecules with the surface. As it follows from the figure, the values for *Ag* and *Ti* are close within the experimental error.



FUGURE 1. Related gas conductivity of the channel M^* for *Ag* and *Ti* atomic clean surface as a function of a gas nature. (The M^* value is standardized on the value of gas conductivity corresponding to completely diffuse interaction of the molecules with the surface).

The completely adsorbed surface is obtained as a result of long exposure of the channel surface preliminary prepared by spraying technique in a residual gas atmosphere of the vacuum chamber (H_2, H_2O, N_2, CO_2) and breaking off fresh spraying of the metal surface before every experiment on the gas conductivity measurement. The results of the free molecular gas conductivity experiments for cylindrical channel with $l/r = 34{,}4$ (*l - length, r - radius of the channel*) for *Ag* and *Ti* atomic clean and for completely adsorbed surface (Me+ads.) in table 1 are submitted.

TABLE 1. The related gas conductivity and the measurement error $M^* \pm M^*$ for different surfaces and gases.				
Surface / gas	**He**	**Ne**	**Ar**	**Kr**
Ag	1.62 ±0.05	1.39 ±0.03	1.23 ±0.04	1.15 ±0.04
Ag+ads.	1.04 ±0.03	1.03 ±0.02	1.0.1 ±0.02	1.00 ±0.02
Ti	1.65 ±0.07	-	1.25 ±0.04	1.14 ±0.05
Ti+ads.	1.04 ±0.02	-	1.02 ±0.02	1.01 ±0.02
Mo+ads.	1.03 ±0.03	1.03 ±0.02	1.02 ±0.02	1.00 ±0.02

On the bases of experimental data on the free molecular gas conductivity for atomic clean and completely adsorbed surface the following basic results can be formulated:

1. Gas conductivity of the channel with atomic clean metal surface is significant higher than one for the surface covered by adsorbate.
2. The sharp expressed dependence of the gas conductivity of the channel with the atomic clean metal surface on a gas nature is observed.
3. It is found that the gas conductivity of the channel with the atomic clean metal surface does not depend on the metal nature (at least for *Ag* and *Ti*).
4. The gas conductivity of the channel with surface covered by adsorbate does not depend significantly on a gas nature and kind of the surface.

INTERPRETATION OF THE EXPERIMENTAL RESULTS

For interpretation the results the theoretical approach created for the molecular beam scattering description is used [4]. According to this approach a gas molecule scattering from a clean metal surface can be related to so called the thermal scattering regime. A thermal vibration of surface atoms at this regime is a dominant factor, which defines of the interaction character. This regime of scattering is characterized by relatively flat effective surface of interaction. At least such surface is seen by an incident atom. That is because a gas - solids interaction takes place at comparatively long distance from the surface. At such distance there is practically no possibility for the gas atom to penetrate into the surface. Gas scattering by physically adsorbed surface is usually related to structural regime of scattering when the effective surface of interaction is rough and interaction takes place at the distance that permits gas atom penetration into the surface. As a result such regime of interaction is characterized by high probability of gas atom capture by the surface and cosinus law of atom reflection after that. This fact at the conditions of this experiment displays in significant decreasing of gas conductivity of the channel with adsorbed surface in comparison with the atomic clean metal surface.

The gas nature influence on the channel conductivity is caused by two reasons. From one side, that is an influence of a gas mass m_0 on the scattering process in thermal regime and this influence grows from *He* to *Kr*. From the other side, that is a result of the potential hole depth D influence which also grows perceptibly from *He* to *Kr* [6]. By this means there is no way to separate the effect of m_0 change from one connected with D in the frame of current experiment as well as using published experimental materials. The effect of D change is caused as a change in a part of incident molecules that can be reflected from the surface after capture. These two effects probably are not independent. Nevertheless, at present time it is accepted that *Ar* and other heavy nobel gases are captured by the metal surfaces more intensive than *He* or *Ne*. As a result the part of the atoms elastic scattered from the surface is significantly larger for light gases than for heavy ones.

As it is shown gas - surface interaction in the thermal regime takes place at the distance when atoms can not feel the nature of surface material. Following to this state one can assume that peculiarity of a gas scattering in the thermal regime is mostly caused by crystal structure characteristics than chemical properties of the surface material. The main characteristic of a crystal metal structure is radius of the surface atom R_S [4]. For instance, $R_s = 1,44$ Å for *Ag, Au, Ti* and majority of metals have R_S values which are close. So that one can expect the same character of scattering by the metal surfaces. This result is confirmed by most of experiments with molecular beams. As an example in the paper [7] almost the same indicatrises of scattering have been obtained for *Ar* atoms on *Ag(111), Pt(111)* and *W(110)* surfaces. At the conditions of our experiment the fact that the gas conductivity does not depend on kind of metal at least for studied surfaces (*Ag* and *Ti*) probably is connected with the same character of noble gas scattering by *Ag* and *Ti* polycrystalline surfaces.

The dominant factor in a structural regime is the roughness of an effective surface of interaction. Such scattering regime is characterized by small radius of interaction (in this case a gas atoms penetrate into the surface) and as a consequence by high probability of gas atom capture by the surface, which then let out atoms according the cosinus law. So called "adsorption regime" take place when the probability of capture (penetration) is equal to unit. It is obvious, that there is no dependence of scattering parameters on the surface and gas nature in the adsorption regime, in this case the gas - solid body interaction completely follows to the diffuse law. In a structural regime such dependence still remains, but is not dominant, as a consequence, the gas conductivity of the channel with surface covered by adsorbate does not depend significantly on a gas nature and kind of the surface.

NUMERICAL SIMULATION OF THE EXPERIMENT

To get the parameters that characterize a gas - solids boundary a numerical simulation of the gas flow through the channel in a free molecular regime has been carried out. As a model of gas scattering by the surface the Maxwell specular-diffuse scheme from the smooth surface has been used. According to the procedure described in the paper [8] the diffuse coefficient is calculated. The results of numerical simulation of the diffuse coefficient for different gases and surfaces are presented in table 2.

TABLE 2. The diffuse coefficient for different gases and surfaces

Surface / gas	He	Ne	Ar	Kr
Ag	0.71	0.80	0.88	0.92
Ti	0.70	-	0.87	0.92
Me+ads.	0.97	0.97	0.99	1.00

The main goal of the numerical simulation is consisted in describing of gas conductivity change as a result of coating degree change during the exposition process. Experimental results obtained earlier [9] are used as a basis for comparison.

Numerical simulation has been carried out in two stages. At the first stage, spreading process of the surface coverage of the channel is simulated. The process is organized with use of oxygen (or other gas) source situated at one side of the channel. At the same stage nonhomogenius coverage degree is calculated as a function of exposition (figure 2 a). At the second stage, with use the test particle Monte Carlo method (TPMC) [10] a gas conductivity of the channel with the surface coverage simulated at the first stage is calculated. It has been assumed, that in the given point of the channel with probability equal of a coverage degree in this point of the channel particle from the adsorbed surface is reflected, hence reflection from an atomic clean surface occurs to probability *(1-)*. Reflection from different surfaces with use of specular-diffuse model with diffuse coefficient following to table 2 is described.

As a result of numerical simulation the dependence of a gas conductivity of the channel on the oxide surface exposure has been obtained (fugure 2 b). This result is in a good agreement with experimental data obtained in the paper [9]. Similar dependence takes place in experiments with the silver surface.

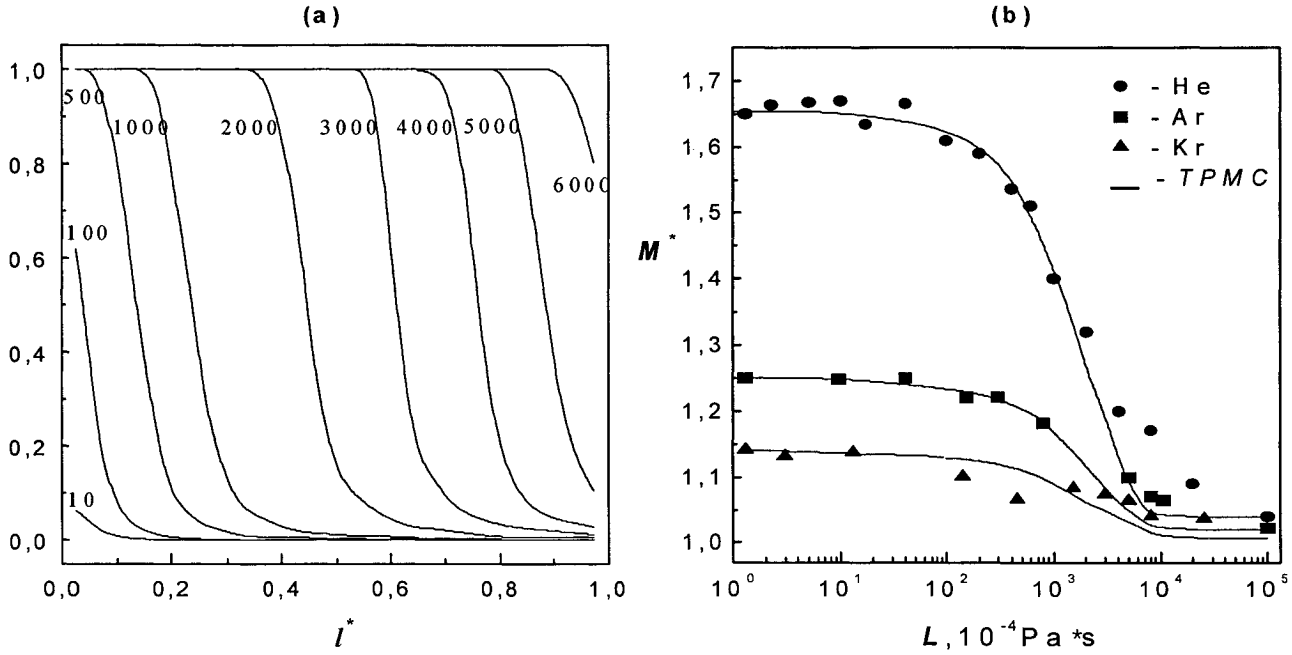


FIGURE 2 (a, b). (a) – The spreading of the oxygen coverage of the cylindrical channel surface from an oxygen source situated at one side of the channel at $l^*=0$. (- a relative degree of coverage, l^* - relative length, the appropriate magnitude of an exposure in Langmuir $1L=10^{-4}$ *Па с* is specified on the curves); **(b)** – the comparison the TPMC calculations with experimental data: related gas conductivity for titanium coated channel as function of surface conditions (oxygen exposure in Langmuir).

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